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17 December 2012

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Office of Naval Research
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From: Eric I. Thorsos, Principal Investigator

Subj: ONR Grant N00014-09-1-0610, "Terahertz Reflection Spectroscopy"

Encl: (1) Final Progress Report for ONR Code 30 SAAET 6.1 Basic Research (with
accompanying SF298)

Please see enclosure (1) for the Final Progress Report for the ONR Code 30 SAAET 6.1 Basic Research Effort, which also includes a list of other articles published over the life of the subject grant. This deliverable constitutes the last documented performance report that will be submitted to the sponsor.

A handwritten signature in blue ink that reads "Eric I. Thorsos".

Eric I. Thorsos

cc: Grant & Contract Administrator, APL-UW
Office of Sponsored Projects
Administrative Contracting Officer, ONRRO Seattle
Naval Research Laboratory Code 5596
Defense Technical Information Center (electronic submission)

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Final Report for ONR CODE 30 SAAET 6.1 Basic Research Effort

1. **Title:** "Terahertz Reflection Spectroscopy For Identification of Explosive Devices"
2. **Prime Offeror:** University of Washington
3. **Subcontractors:** None
4. **Period of Performance:** March 1, 2009 – August 30, 2012
5. **Submitted by:** Eric I. Thorsos, eit@apl.washington.edu, 206-543-1369
6. **Business Contact:** Jody Khan-Hauge, jkhaug@apl.washington.edu, 206-543-7814
7. **Background/Scope of Effort:** The physics of interaction between key materials and electromagnetic radiation at terahertz (THz) frequencies of 10^{12} to 10^{13} Hz has been considered promising, on two counts, for the detection of concealed explosives. First, characteristic absorption features in reflection spectra of explosives may provide a basis for discrimination from other materials. Second, common obscuring materials (e.g., cellulose, plastic) are translucent to THz radiation, so detection and imaging of spectral signatures inside containers have considerable potential. However, realization of this potential is both mediated by, and complicated by, effects of scattering. Scattering from the granular structures and rough surfaces of actual explosives may distort or obscure the spectral features. This is a general issue with all THz techniques for imaging and detection of a strongly scattering target. It is crucial to understand the volume and rough surface scattering processes, to understand the interaction between scattering and spectral absorption, and to develop new theoretical models and new technology to extract the spectral absorption information in the presence of strong scattering. On the other hand, scattering may make spectral signature more easily detectable, because diffuse scattering can send the scattered energy over a wide range of backscattering angles, much more convenient for detection than the case of specular reflection from a planar target, where the geometry would need to be tightly controlled to permit detection of spectral signatures.
8. **Summary/Abstract:** The research conducted under this project was a joint effort between Applied Physics Laboratory, University of Washington (APL-UW), and Portland State University (PSU). The focus at APL-UW was on 1) developing an experimental setup for variable source and detector angles and variable linear polarization, and then implementing it on a commercial THz spectroscopic system at PSU, 2) expanding the current THz time domain spectroscopic system at APL-UW to cover broader bandwidth by developing an air plasma THz source, and 3) developing analysis techniques that allow spectroscopic signatures to be recovered in the presence of appreciable background noise, a situation that can be expected when volume and rough surface scattering processes are important. The long-term goal of this project has been to lay the groundwork for an integrated spectral and imaging methodology on which to base technology development.

9. Technical Contents and Accomplishments

9.1. *Implementation of an automated measurement system*

The task of the first program year was to implement automated variable transmitter and receiver angles and variable linear polarization measurements on a commercial THz spectroscopic system at Prof. Lisa Zurk's laboratory at Portland State University. Prof. Zurk was a co-PI of our previous project in the SAAET program, and although she now leads a separate SAAET project, her project and our project have been collaborative in nature with similar goals.

When a THz wave is scattered on a rough surface, the incident THz wave is re-distributed over a wide range of angles and the state of polarization of the scattered wave can be different from that of the incident wave. It is necessary to perform THz scattering measurements at various angles and polarizations to accurately describe the scattering properties of a test sample. PSU acquired a commercial THz spectroscopy system made by Picometrix. It is a fiber laser based system that offers great flexibility and allows the transmitter and receiver heads to be moved freely and be arranged with any angles of incidence and reflection with respect to the test sample. Because scattering is a stochastic process in a random medium or at a randomly rough surface, a large amount of data is needed to develop and validate statistical models. To accomplish this requires a system of computer controlled motion stages in precisely coordinated movement.

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The system of motion stages was designed and constructed at APL-UW (Figure 1). It consists of a stack of two rotation stages to rotate the sample and the receiver (the transmitter is stationary and the change of angle of incidence is achieved by rotating the sample), an X-Y translation stage for in-plane motion of the sample for different realizations of the scattering measurement, and two rotation stages to rotate the transmitter and receiver heads, respectively. After it was assembled and tested it was transferred to PSU and installed with the Picometrix transmitter and receiver heads to form an automated THz scattering measurement system. The APL-UW team trained the PSU team on how to interface the system with computers and LabView software, and how to operate the system. The APL team also provided technical support throughout the duration of the project.

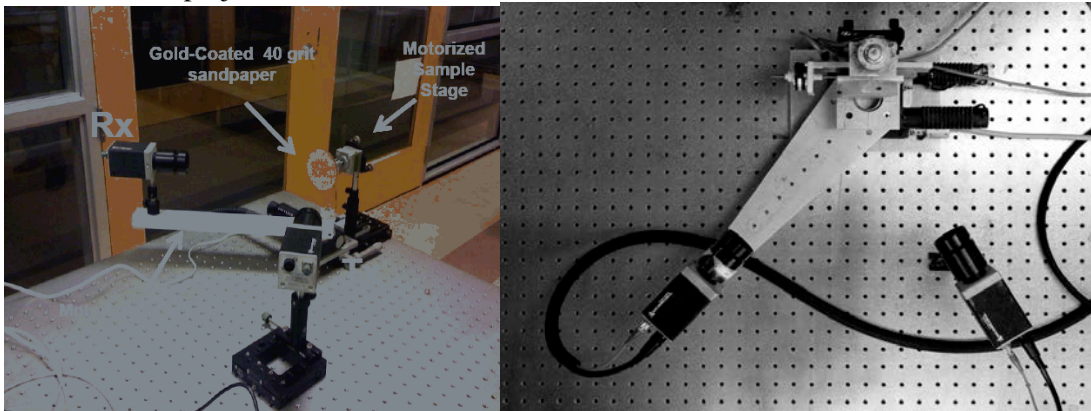


Figure 1. A view of the automated motion stages installed on the PSU THz spectroscopy system.

The automation of the measurement system enabled the two groups to do extensive and systematic studies of THz scattering. Without the automation system, setting up different measurement configurations and collecting large amounts of data had to be done manually and was prone to human error. The system proved to be highly useful for research during the subsequent program years.

9.2. Development broadband air plasma THz source

The approach taken to develop a broader band THz source was to utilize an air plasma for the source. The nonlinear process by which the third order optical susceptibility, $\chi^{(3)}$, contributes to the generation of THz radiation is known as four-wave mixing. It was first shown by Cook and Hochstrasser [1] in 2000 that the air plasma generated by ionization of air molecules, when a femto-second laser beam is focused in the ambient air, exhibits a very strong third order susceptibility tensor, which can be exploited to efficiently generate intense coherent THz radiation. The process uses the nonlinear interactions of the fundamental wavelength photon, which also produces the air plasma, with its second harmonic photons generated in a BBO crystal. We have implemented a four-wave mixing THz source in our THz-TDS setup. Figure 2 shows photographs of the air plasma size and intensity generated using our amplified femtosecond laser beam with average power of 800 mW, center wavelength of 790 nm, and pulse length of 50 nm or 45 fs, produced by focusing through lenses with various focal lengths. Note that the camera exposure time has been significantly increased from the strongest lens to the case with the longest focal length in order to resolve and fully capture the ionized region in the ambient air.

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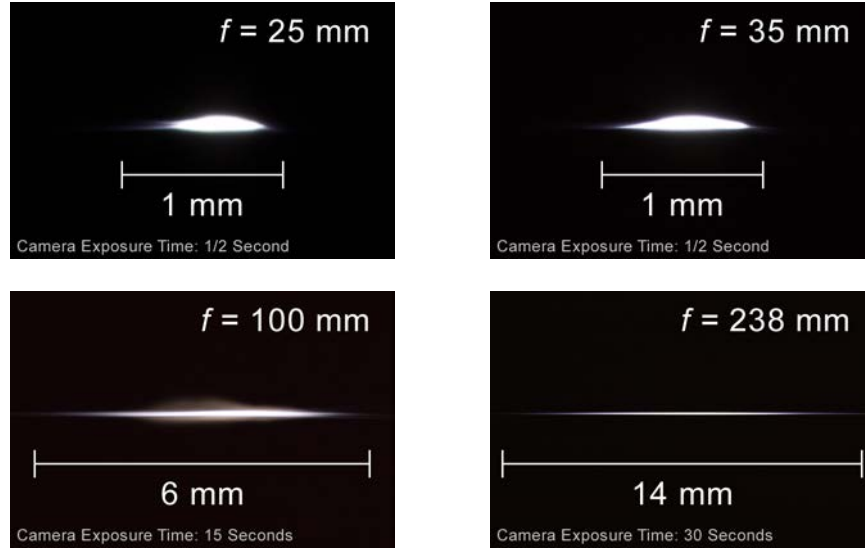


Figure 2. Effect of varying the focal length f on the size and intensity of the air plasma produced, using the femtosecond amplified laser system at UW-APL.

It has been shown experimentally that tighter focusing produces a more intense air plasma; however, for physical considerations such as the phase matching conditions, the length of interaction between the fundamental and its second harmonic, the profile of the air plasma region, as well as the convergence angle of the THz radiation, longer focusing results in stronger terahertz emission [2-6]. Figure 3 shows the higher power and broader bandwidth of the terahertz waves generated using this technique in a N_2 purged air plasma source vs. optical rectification in ZnTe for comparison. We have also explored using various EO crystals to extend the detection of bandwidth of our system to support future experiments. Figure 3 also shows that a broader bandwidth can be achieved using a GaP detector compared to ZnTe crystals. This experimental capability would allow measurements of incoherent THz spectroscopy from rough surface substances to be expanded to much higher THz frequencies.

9.3. Implementation of multi resolution analysis for identifying spectroscopic features

When attempting to identify spectroscopic features in a THz signal scattered from a target with a rough surface or with volume heterogeneity, the signal is likely to be embedded in background noise produced in the scattering process [7]. This can be illustrated by observing how the strength of a spectral feature decreases as the roughness on a test sample increases. We chose as our test material α -lactose monohydrate, whose absorption signatures at 0.54, 1.20, and 1.38 THz are well known. Controlled levels of surface roughness were inscribed in the pellets from three different grades of sandpaper sheets, designated as 400, 150, and P180 grit. We also measured the roughness parameters of the sandpaper independently to support theoretical modeling. We then used our THz time-domain spectrometer (optical rectification in ZnTe) to measure scattered radiation in the specular direction for separate patches on the rough surface, from which we computed the mean power, individual incoherent scattered powers (power for a single realization minus the mean coherent power), and the mean incoherent scattered power, all as functions of THz frequency. A sample of results is shown in Figure 4, where the roughness increases as the grit size decreases. Figure 4 shows the Fourier amplitude of the reflected THz field as a function of frequency, and the lactose spectroscopic feature of primary interest is at 0.54 THz.

Important spectroscopic features can be made more easily visible by plotting the first derivative of the spectral amplitude, as shown in Figure 5 where the frequency range as been

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reduced and centered on the spectral feature of interest. Note that as the roughness increases, the visibility of the absorption feature decreases and is effectively lost for the two roughest cases. We also found the frequency dependence of coherent power to be in close agreement with simple theoretical modeling based on the Kirchhoff approximation (inset). This information was used to detrend the spectral amplitudes prior to the wavelet analysis to be described shortly.

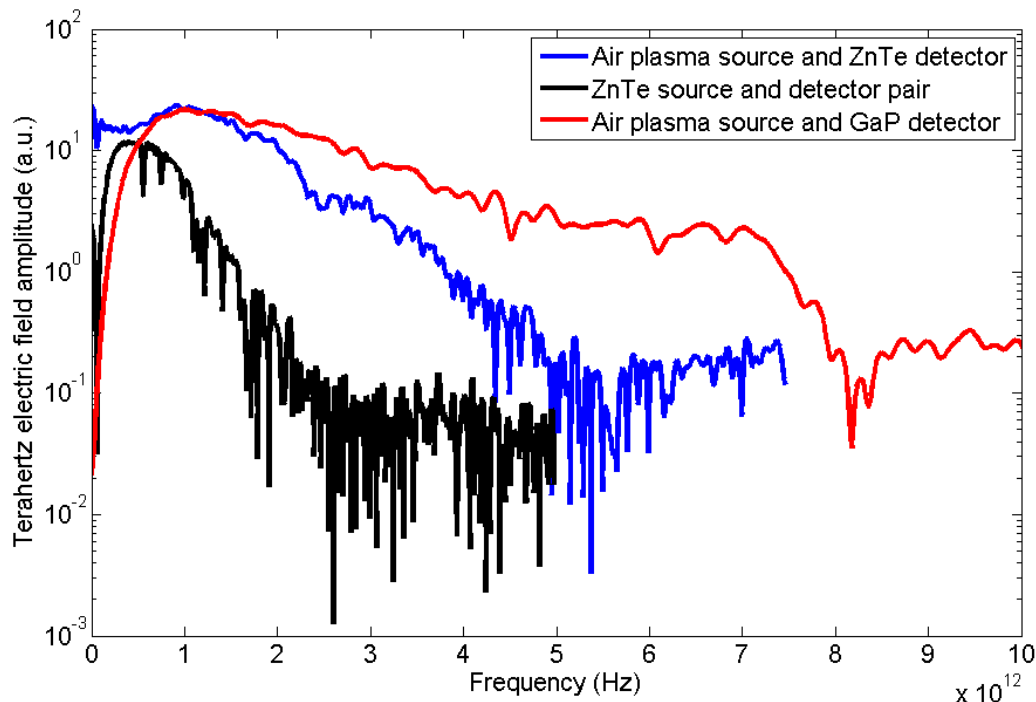


Figure 3. A comparison between the spectral content of the terahertz field generated using both the four-wave mixing process in an air plasma and optical rectification in a ZnTe crystal. The terahertz fields are detected by the EO-sampling method using both a ZnTe and GaP crystal.

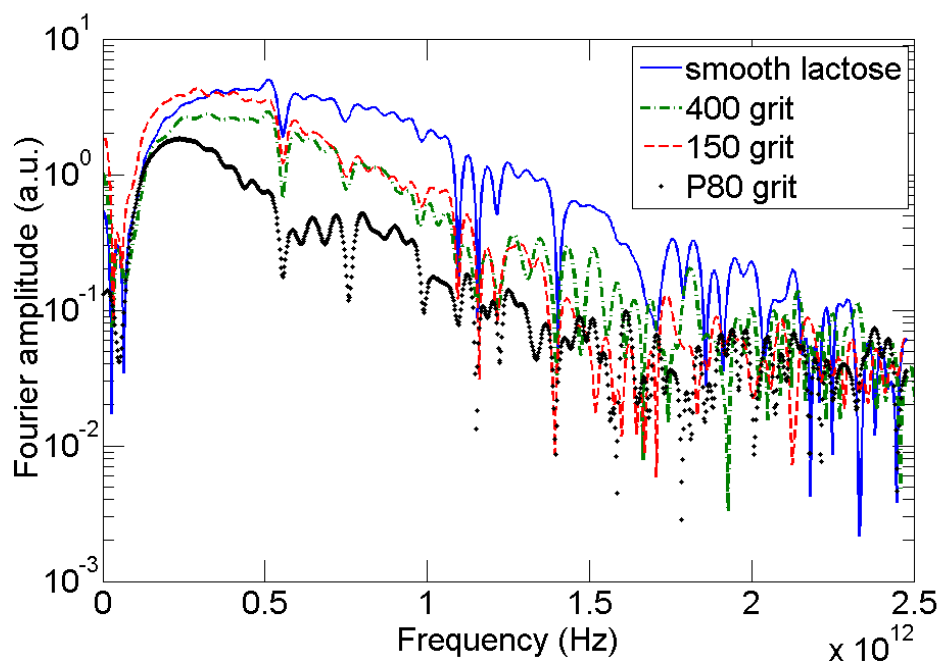


Figure 4. Fourier amplitude of the reflected THz field for smooth and rough lactose pellets.

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The result in Figure 5 illustrates the need for signal processing techniques that can detect the presence of spectral features that might be obscured by fluctuations in the background level (which we refer to as noise). Motivated by this need, we have developed a method using wavelet decomposition of the frequency-domain signal that successfully recovers the location of the spectral feature (resonance) even in the roughest case shown in Figure 5 (P80 grit). Wavelet methods are particularly suitable for analysis of data series with localized features, whose locations are not known *a priori*, as opposed to Fourier-based spectral analysis techniques that work best with stationary functions. The wavelet method works essentially by separating variations in the frequency-domain signal into a sequence of scale sizes (in this case frequency scales) with the effect of separating out the fluctuations in the background caused by incoherent scattering from the specific scale of the resonance, thus providing a statistically sound basis for detection of the resonance even when it is obscured by incoherent scattering effects.

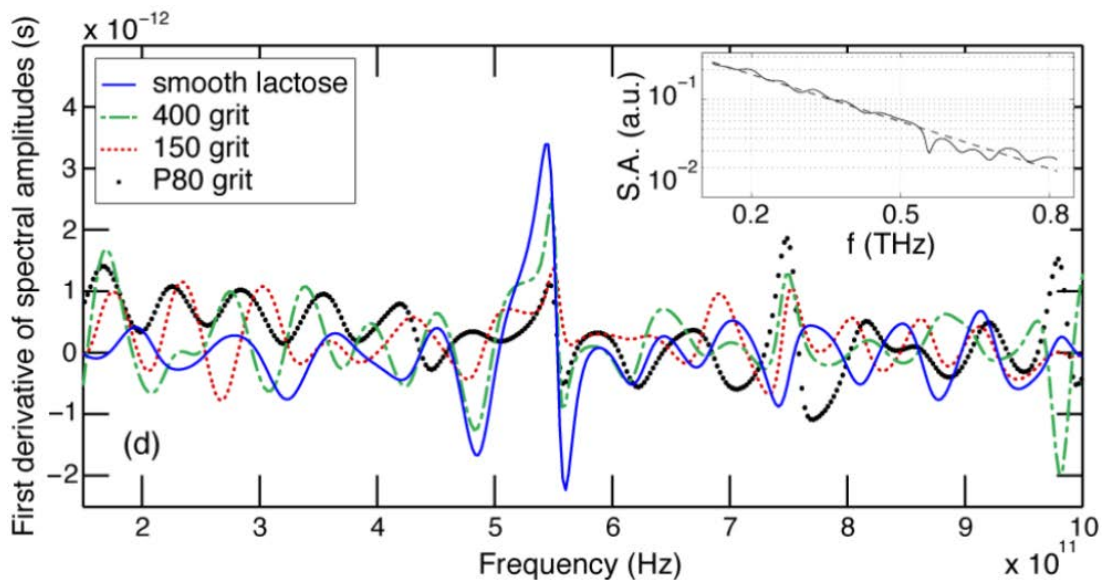


Figure 5. First derivative of reflection spectral amplitude of lactose pellets from Figure 4.

A detailed discussion of wavelet methods in general and the particular choices made for application to analyzing terahertz reflection spectra would require an extensive treatment, even to clarify the technical terms used in describing the analysis. Such an extensive treatment will not be included here since the details are available in the literature. A very useful treatment on wavelet methods in general can be found in the monograph by Percival and Walden [8]. Details on our analysis are also available [9-11].

An illustration of how the wavelet method can separate the variations in the frequency-domain signal into a sequence of frequency scale sizes is shown in Figure 6, where the original detrended spectral data series from the roughest case in Figures 4 and 5 (roughness obtained with the P80 grit) is shown at the top and labeled X. The vertically offset plots below result from a wavelet decomposition of data series X into a sequence of components ranging from the largest frequency scale size at the top to the smallest at the bottom. When the eight offset plots \tilde{D}_1 to \tilde{D}_8 are added together they reconstruct the original data series X. The absorption feature of lactose near 0.54 THz is evident in \tilde{D}_2 and \tilde{D}_3 .

In Figure 6 all of the offset plots have the same vertical scale, causing the signature of the absorption feature at 0.54 THz to become small for the shortest scale sizes, though the signal to noise ratio remains good. Thus, expanding the vertical scale for the shortest scale sizes is

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beneficial. This is shown in Figure 7 for data obtained with the air plasma source illuminating roughened lactose (P80 grit).

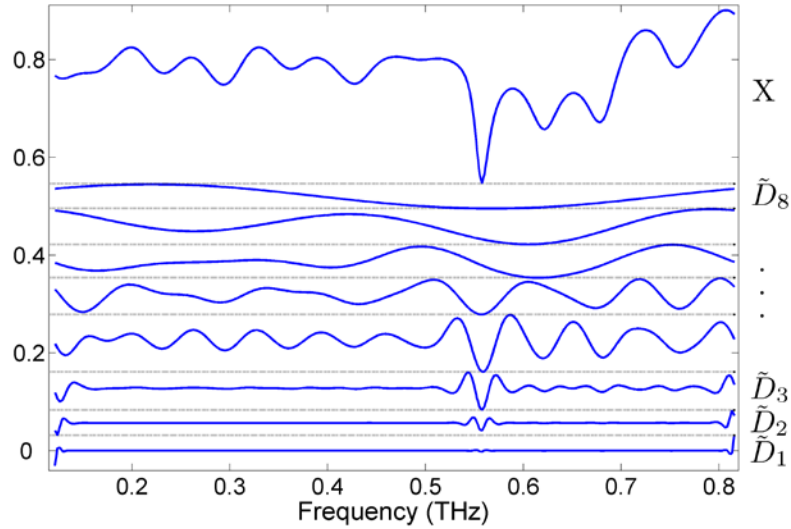


Figure 6. Separation of the original rough lactose (P80 grit) detrended spectral data series X into a sequence of spectral scale sizes using the wavelet method. Vertically offset plots show components of the original data series ranging from large to small scale size (top to bottom). The absorption resonance of lactose near 0.54 THz is evident in \tilde{D}_2 and \tilde{D}_3 . The vertical axis is dimensionless.

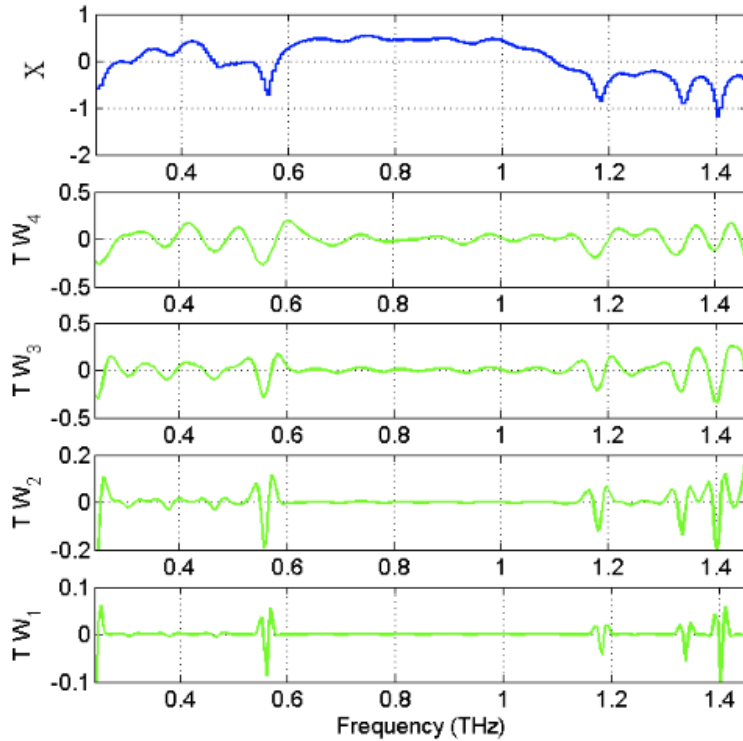


Figure 7. Separation of the original rough lactose (P80 grit) detrended spectral data series X into a sequence of spectral scale sizes using the wavelet method. The air plasma source was used for this case, and only the four smallest scale components are shown.

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The data series components shown in Figure 7 correspond to the four lowest shown in Figure 6, though the notation labeling the components has been changed. It is evident that for shortest spectral amplitude scale (\tilde{D}_1 in Figure 6, TW_1 in Figure 7) the absorption resonance feature of lactose near 0.54 THz has the highest signal to noise, which would facilitate an automated detection algorithm. Additional absorption features also appear in the region above 1 THz for this air plasma source generated data series.

Another issue that would need to be addressed for an automated detection algorithm is the detrending step that was used to remove the main decrease in signal level as the frequency increases, shown in Figure 4. The detrending operation in that case was performed based on the frequency dependence of the coherent reflection loss from a rough surface, and then numerically fitting that trend to the data as shown in the inset in Figure 5. It would be much better, however, to have a universal detrending algorithm that does not depend on fitting a theoretical scattering prediction to the data. It happens that the wavelet approach being used imposes circular boundary conditions on the data series, which is one reason that an initial detrending step is made before the wavelet-based decomposition is performed. An approach was developed to avoid the detrending step altogether by using the original data series as the first quarter of an artificial periodic extension such that the resultant extended data series would repeat itself by the completion of the full period. Figure 8 illustrates this procedure.

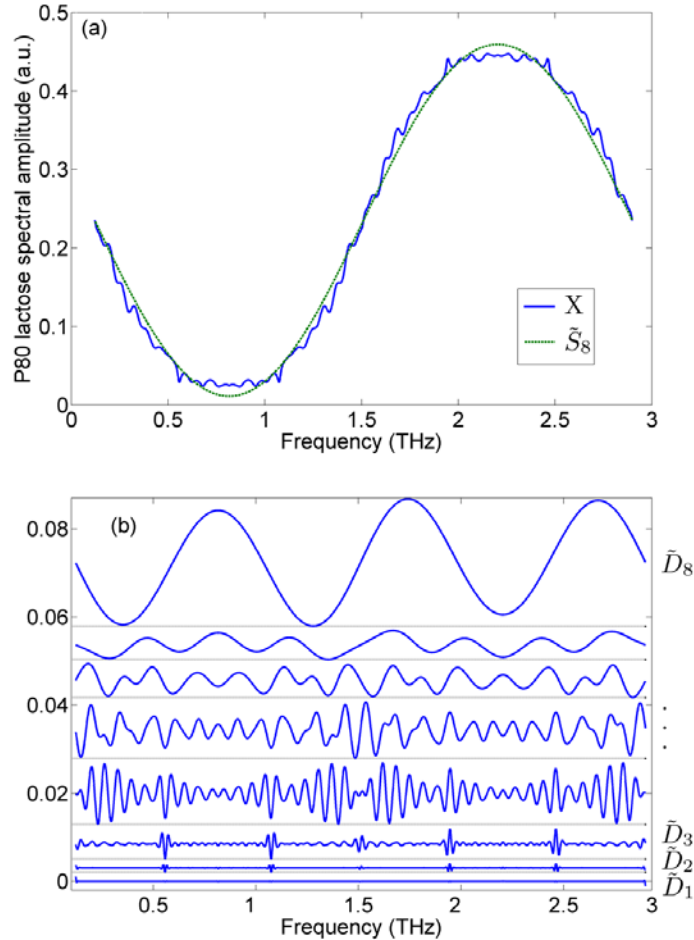


Figure 8. (a) A periodic extension of the P80 lactose spectral amplitude (solid line) along with the so-called smooth part (dotted line) which carries the dominant large-scale structure. (b) The wavelet decomposition of the data series, similar to Figure 6, after removal of the smooth part.

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Figure 8(a) shows the periodic extension of the original P80 grit spectral amplitude along with a “smooth part” that also is obtained as part of the wavelet processing scheme. After its removal, the decomposition into a series of spectral scale sizes proceeds as before, and the spectral absorption feature along with its periodic extension is visible in the \tilde{D}_2 and \tilde{D}_3 components. If the vertical axis were expanded as in Figure 7, then the absorption feature would be just as visible in \tilde{D}_1 as well. Following this procedure an automated algorithm could easily be implemented for detecting specific spectral features.

Another issue that is worth understanding is the extent to which the wavelet analysis can distinguish closely spaced spectral features. To study this topic, a realistic model for an absorption feature was added to a measured spectral amplitude obtained from smooth lactose. The result is a spectral amplitude with two absorption features, one at the native 0.54 THz of α -lactose, the second at a frequency that was varied in steps from 0.5 to 0.6 THz. The results are shown in Figures 9-13. As with Figure 7, only the four smallest-scale components obtained from the wavelet analysis are shown, and the components are also labeled as in that figure. It can be seen that as the second peak moves from 0.5 to 0.6 THz, except for when it is located at 0.55 THz, the existence of two separate features can be clearly identified. When the twin feature is placed at 0.55 THz in Figure 11, very close to the native one at 0.54 THz, it is still possible to observe that the structure is more complex than obtained from a single absorption resonance. Therefore, the possibility remains that with suitable modeling input, an automatic algorithm could be produced that would recognize this special situation and flag it for more detailed analysis.

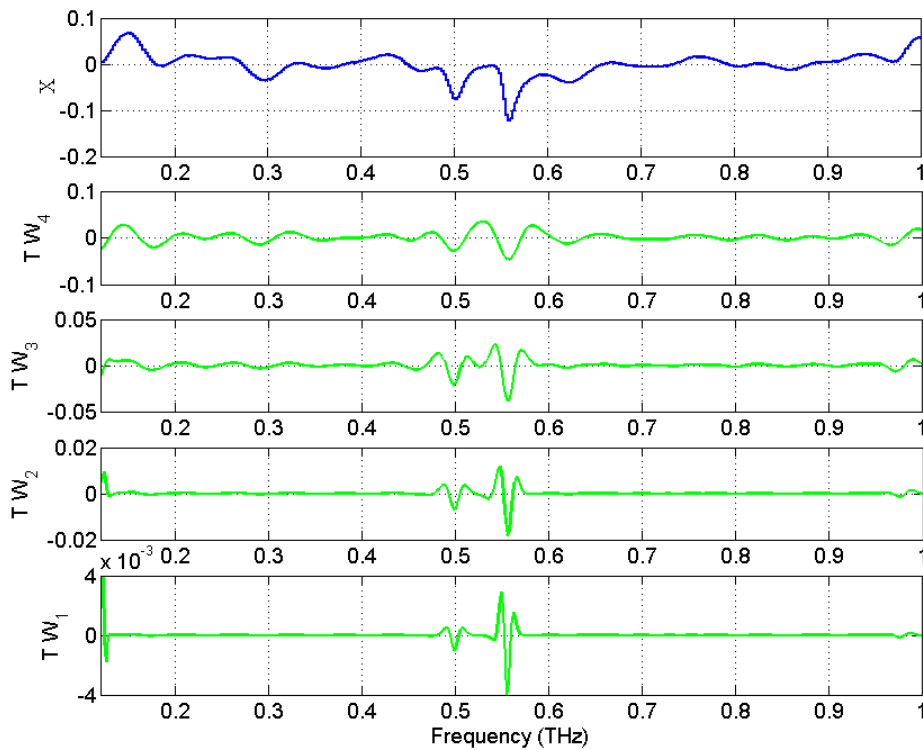


Figure 9. The THz reflection spectra of an artificial material with the additional simulated twin absorption feature of lactose at 0.50 THz.

In summary, the wavelet method that has been developed shows considerable promise in identifying spectral features that may be embedded in background noise. A patent application has been filed through the University of Washington Office of Technology Transfer on this method, which likely has application well beyond THz reflection spectroscopy.

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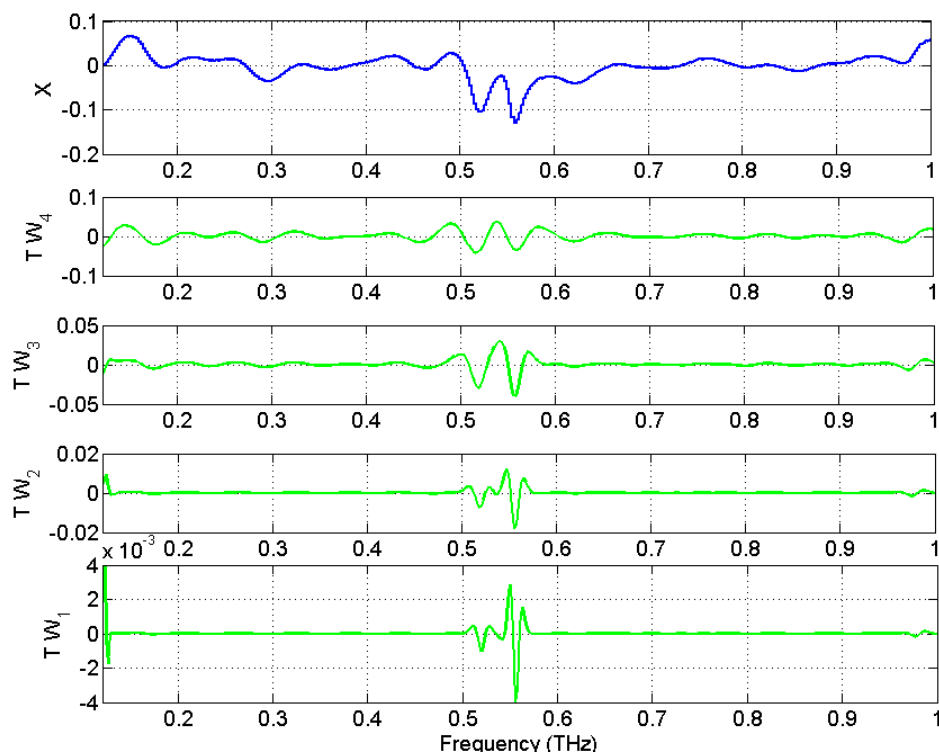


Figure 10. The THz reflection spectra of an artificial material with the additional simulated twin absorption feature of lactose at 0.52 THz.

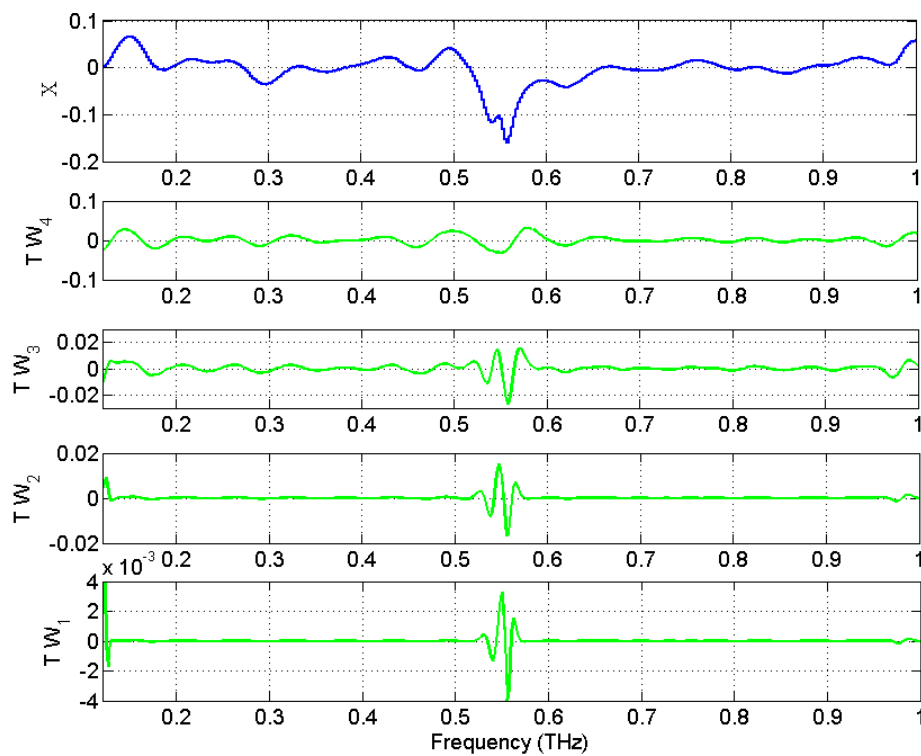


Figure 11. The THz reflection spectra of an artificial material with the additional simulated twin absorption feature of lactose at 0.55 THz.

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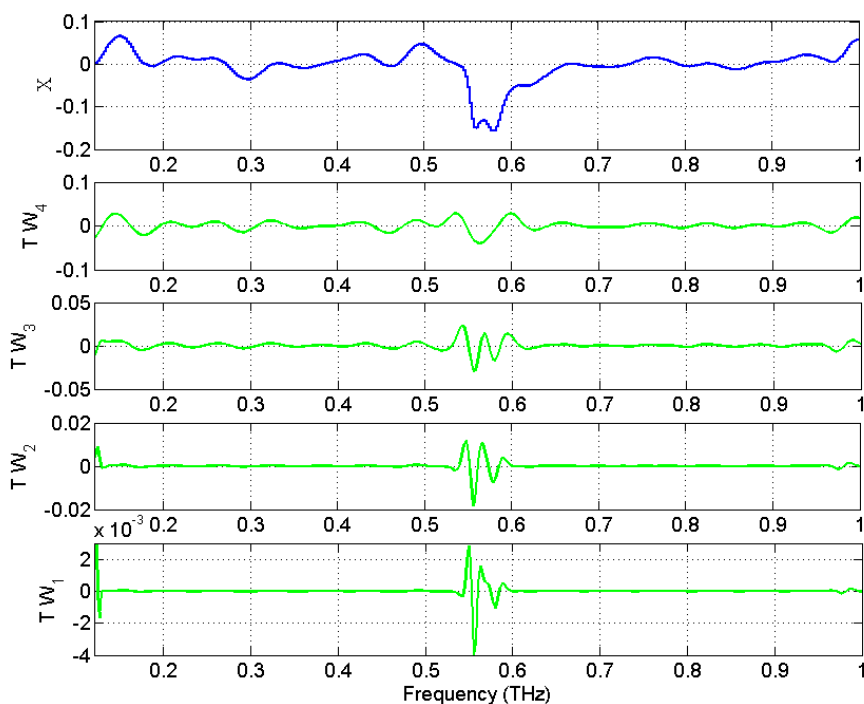


Figure 12. The THz reflection spectra of an artificial material with the additional simulated twin absorption feature of lactose at 0.58 THz.

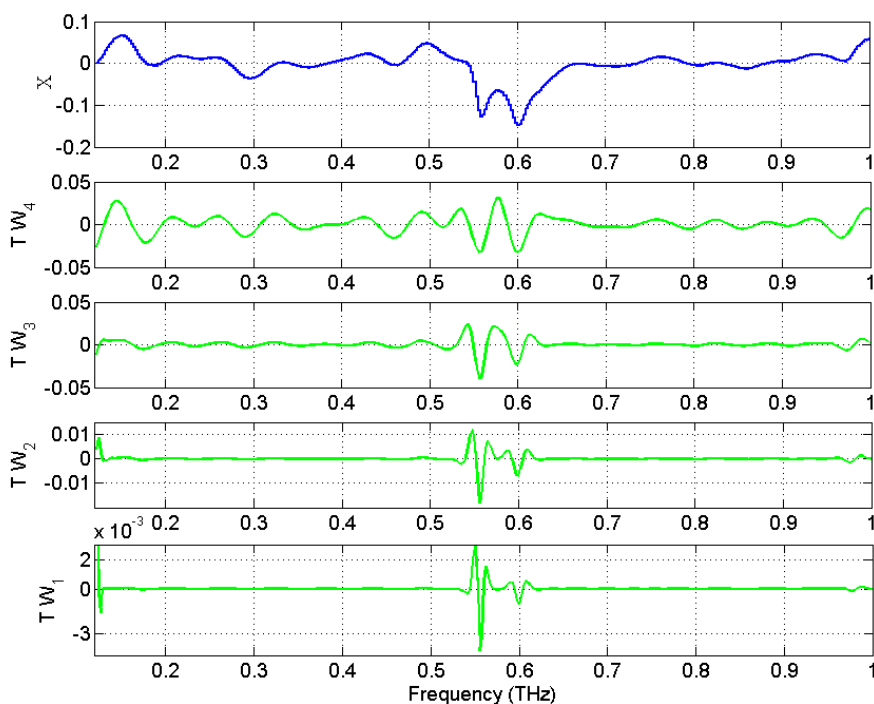


Figure 13. The THz reflection spectra of an artificial material with the additional simulated twin absorption feature of lactose at 0.60 THz.

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10. Deliverables, Personnel, Awards, Publications, Presentations, and Patents

Deliverables	Date
“Terahertz spectroscopy of rough surface targets,” M. H. Arbab, D. P. Winebrenner, A. Chen, and E. I. Thorsos, United States Patent Application Publication, US 2012/0191371 A1, July 26, 2012.	January 20, 2010
Final Technical Report with SF298	December 2012

Table 1. Deliverables

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	Total #	Name	Organization
PI	1	Eric I. Thorsos	University of Washington
Co-PIs	1	Antao Chen	University of Washington
Graduate Students / Research Assistants	1	M. Hassan Arbab	University of Washington

Table 2. Personnel Information

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	Name	Award (Year Received)	Prize (Year Received)	Recognition (Year Received)
PI	Eric I. Thorsos	Applied Physics Laboratory Director's Award (2010)		

Table 3. Awards/Prizes/Recognitions

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Name	Publication (Date)	Conference Presentation (Date)	Patent (Date)
M. H. Arbab, D. P. Winebrenner, E. I. Thorsos, and A. Chen	<u>"Application of wavelet transforms in terahertz spectroscopy of rough surface targets," <i>Proc. SPIE</i> vol. 7601, pp. 760106:1-7 (2010).</u>	Presented in SPIE Photonics West Conference, January 2010, San Francisco, CA	
M. H. Arbab, D. P. Winebrenner, E. I. Thorsos, and A. Chen	"Retrieval of terahertz spectroscopic signatures in the presence of rough surface scattering using wavelet methods," <i>Appl. Phys. Lett.</i> 97 , 181903 (2010).		
M. H. Arbab, T. C. Dickey, D. P. Winebrenner, A. Chen, and P. D. Mourad	"Characterization of burn injuries using terahertz time-domain spectroscopy," <i>Proc. SPIE</i> 7890 , 78900Q, 2011	Presented in SPIE Photonics West Conference, January 2011, San Francisco, CA	
M. H. Arbab, T. C. Dickey, D. P. Winebrenner, A. Chen, M. B. Klein, and P. D. Mourad	"Terahertz reflectometry of burn wounds in a rat model," <i>Biomedical Optics Express</i> 2 , 2339-2347, 2011.		
M. H. Arbab	Terahertz spectroscopy for chemical detection and burn characterization," Ph.D. thesis, University of Washington (2012).		
M. H. Arbab, D. P. Winebrenner, A. Chen, and E. I. Thorsos			"Terahertz spectroscopy of rough surface targets," United States Patent Application Publication, US 2012/0191371 A1, July 26, 2012.
M. H. Arbab, D. P. Winebrenner, T. C. Dickey, A. Chen, M. B. Klein, and P. D. Mourad,	"A Non-invasive Terahertz Assessment of 2 nd and 3 rd Degree Burn Wounds," <i>Proc. CLEO</i> , San Jose, CA, 2012	Presented in CLEO Conference, May 2012 San Jose, CA.	

Table 4. Publications, Conference Presentations and Patents

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11. References: (In support of Sections 7-9 above.)

- [1] D. J. Cook and R. M. Hochstrasser, "Intense terahertz pulses by four-wave rectification in air," *Opt. Lett.* **25**, 1210-1212 (2000).
- [2] J. Dai et al., "Detection of broadband terahertz waves with a laser-induced plasma in gases," *Phys. Rev. Lett.* **97**, 103903 (2006).
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- [4] H. Zhong et al., "Terahertz emission profile from laser-induced air plasma," *Appl. Phys. Lett.* **88**, 261103 (2006).
- [5] T. Bartel et al., "Generation of single-cycle THz transients with high electric-field amplitudes," *Opt. Lett.*, **30**, 2805-2807 (2005).
- [6] M. Kress et al., "Terahertz-pulse generation by photoionization of air with laser pulses composed of both fundamental and second-harmonic waves," *Opt. Lett.* **29**, 1120-1122 (2004).
- [7] M. H. Arbab et al., "Effect of surface scattering on terahertz time domain spectroscopy of chemicals," *Proc. SPIE* **6893**, 68930C (2008).
- [8] D. B. Percival and A. T. Walden, *Wavelet Methods for Time Series Analysis*. New York, Cambridge Univ. Press, 2008.
- [9] M. H. Arbab, *et al.*, "Application of wavelet transforms in terahertz spectroscopy of rough surface targets," *Proc. SPIE* **7601**, 760106-7 (2010).
- [10] M. H. Arbab, *et al.*, "Retrieval of terahertz spectroscopic signatures in the presence of rough surface scattering using wavelet methods," *Appl. Phys. Lett.* **97**, 181903 (2010).
- [11] M. H. Arbab, "Terahertz spectroscopy for chemical detection and burn characterization," Ph.D. thesis, University of Washington (2012).

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14. ABSTRACT The research conducted under this project was a joint effort between Applied Physics Laboratory, University of Washington (APL-UW), and Portland State University (PSU). The focus at APL-UW was on 1) developing an experimental setup for variable source and detector angles and variable linear polarization, and then implementing it on a commercial THz spectroscopic system at PSU, 2) expanding the current THz time domain spectroscopic system at APL-UW to cover broader bandwidth by developing an air plasma THz source, and 3) developing analysis techniques that allow spectroscopic signatures to be recovered in the presence of appreciable background noise, a situation that can be expected when volume and rough surface scattering processes are important. The long-term goal of this project has been to lay the groundwork for an integrated spectral and imaging methodology on which to base technology development. The approach toward equilibrium, as measured $\ln(1+h) = -\ln(1-h) = h + \frac{h^2}{2} + \frac{h^3}{3} + \dots$ is exponential: $2(t) = 12 \exp(-\frac{1}{2} t \ln(1+h)) + 1 + O(h^2)$. This provides a ical chaos.					
15. SUBJECT TERMS spectroscopy, variable linear polarization, spectroscopic signature recovery					
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